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Size Controllable DNA Nanoribbons Assembled from Three Types of Reusable Brick Single-Strand DNA Tiles

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Abstract

Precise control of nanostructure is a significant goal shared by super molecular chemistry, nanotechnology and materials science. In DNA nanotechnology, methods of constructing desired DNA nanostructures by programmable DNA strands have been studied heavily and become a promising research branch, but developing universal and low cost (in sense of using less types of DNA strands) method remains a challenge. In this work, we propose a novel approach to assemble size controllable DNA nanoribbons with three types of reusable brick SSTs (single-strand DNA tiles), where the control to ribbon size is achieved by regulating the concentration ratio between manipulative strands and packed single stranded DNA tiles. In our method, three types of brick SSTs are sufficient in assembling DNA nanoribbons of different sizes, which is much less the number of types of unique tile programmable assembling strategy, thus achieving a universal and low cost method. The assembled DNA nanoribbons are observed and analyzed by atomic force microscopy (AFM). Experimental observations strongly suggest the feasibility and reliability of our method.

Keywords

DNA Nanotechnology, DNA nanoribbon, DNA nanotube, Controllable shape, Low cost strategy

1. Introduction

DNA (deoxyribonucleic acid), well known as the carrier of genetic information in cell, has emerged recently as effective nano elements for constructing nanostructures in recent years, such as DNA nanoribbons, tubes and lattices, as well as for developing nano scale materials [1-8]. DNA nano materials provide a great potential for bottom-up nanofabrication and function structures with prescribed features [9-19]. Plenty of approaches have been proposed to produce DNA nano structures with controllable patterns, shapes and lengths. Till now, there are three mainly investigated methods to produce DNA nano structures: self-assembly of DNA tiles [20-22], DNA origami [23-26] and single-stranded tiles (SSTs) based on approaches [27, 28]. Study on using DNA molecules to produce programmable DNA nano structures becomes a hot and promising research branch in DNA nanotechnology

In 2005, Yan *et al.* proposed an innovative strategy of self-assembly symmetric finite-size DNA nanoarrays [21]. After that, Rothemund *et al.* reported a so called "easy" way for folding a 7-kilobase single-stranded scaffold into a desired shape by using over 200 short oligonucleotide "staple strands" [23]. Andersen *et al.* improved the method by designing an addressable DNA box to fold six DNA origami sheets and then patch the box by staple strands [26]. In 2012, Peng *et al.* proposed the method of using SSTs (single-stranded DNA tiles) to develop a molecular canvas consisting of 310 pixels, where each constituent SST strand acted as a pixel and the pattern can be controlled by keeping and annealing strands being covered by the target shape [27]. Following the research line, Peng *et al.* extended the SST method to construct prescribed three-dimensional (3D) shapes by simply selecting subsets from a common 3D cuboid "molecular canvas" with dimensions of 10 by 10 by 10 voxels [28].

The significant methods reported in [21-28] can produce different programmable DNA nanostructures with controllable patterns, shapes and lengths, but usually a big or even a large number of uniquely addressable DNA strands are needed, which make the methods cost and sometimes time consuming. Meanwhile, large numbers of DNA strands will pose challenges in designing specific domains with minimum similarity and make DNA self-assembly more error-prone. From these points of view, universal and low cost (in sense of using less types of DNA strands) method is needed, but remaining a challenge in DNA nanotechnology.

In this work, we propose a novel approach to assemble size controllable DNA nanoribbons with only using three reusable brick SSTs. It starts by assembling DNA nanotubes with the brick SSTs, from which DNA nanoribbons (in rectangle) can be obtained by adding manipulative DNA strands. (Note that, the length of DNA nanotubes can be controlled by the length of SSTs.) In our method, except for brick

SSTs, packed strands and manipulative strands are also designed, where packed strands are paved periodically in the plane by means of hybridizing with other packed strands; manipulative strands for bottom-sealing are single strands and will be applied to control the self-assembly scale of packed strands. By regulating the ratio between concentrations of manipulative strands for sides-sealing and packed strands, DNA nanoribbons with different sizes can be assembled. In the assembling process, three types of brick SSTs are sufficient to generate DNA nanoribbons of different sizes, thus achieving a universal and low cost method.

The assembled DNA nanoribbons are observed and analyzed by atomic force microscopy (AFM). With the repeated AFM scanning, we have not found the tube opening as DNA tile tube opened under AFM before. This is due to the fact that longer sticky ends of single strand tile can give more Thermodynamic stability of this tube structure. Experimental observations and AFM image analysis suggested the feasibility and reliability of our method.

2. Design and Modeling

In the method, three brick SSTs, labelled by S1, S2 and S3, are used to assemble DNA nanoribbons with different sizes. In general, each SST is composed of 4 domains (represented by colored line segments as shown in Figure 1).

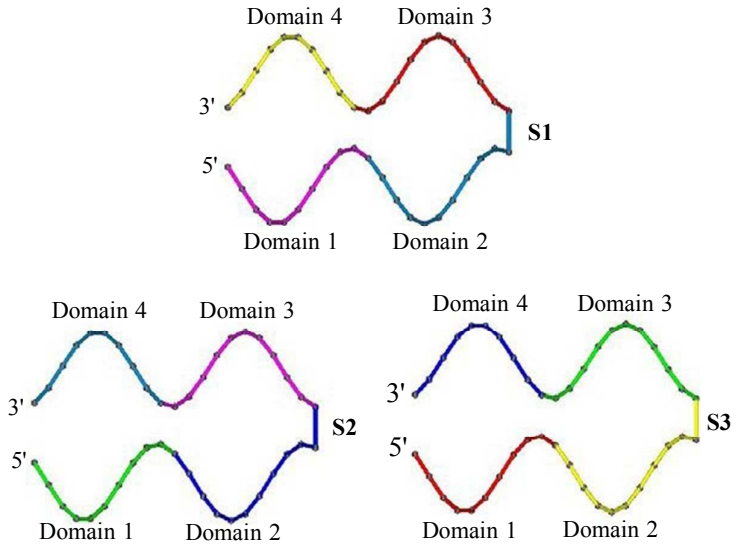


Figure 1. Brick SSTs S1, S2 and S3, where colored lines represent modular domains

In SSTs shown in Figure 1, domains with the same color are complementary according to Watson-Crick base pairing principle and anti-parallel with each other to form double DNA strands. The following pairs of domains are with complete complementary sequences: domain 1 of S1 and domain 3 of S2, domain 2 of S1 and

domain 4 of S2, domain 3 of S1 and domain 1 of S3, domain 4 of S1 and domain 2 of S3, domain 1 of S2 and domain 3 of S3, domain 2 of S1 and domain 4 of S3. With the help of complementary domains, specific two SSTs can form helix structure, including S1&S2, S2&S3, S3&S1. Every helix structure is composed of length 21 nucleotides double-stranded DNA (ds DNA) and two sticky ends of single strands. For example, in S1&S2 helix structure, two sticky ends are domains 3-4 of S1 (3'-end) and domains 1-2 of S2 (5'-end). The cyclization of helices can be finally facilitated by the accumulation of intrinsic curvature of every SST, see Figure 2(a). Note that, if no additional strand is added, it will form DNA nanotubes, where the length of helix is exactly the length of nanotube (see Figure 2(b)). This is the product of the repeating tilling of the same dsDNA segments. The nanotubes constructed by brick SSTs have the circumference $w \approx 12nm$.

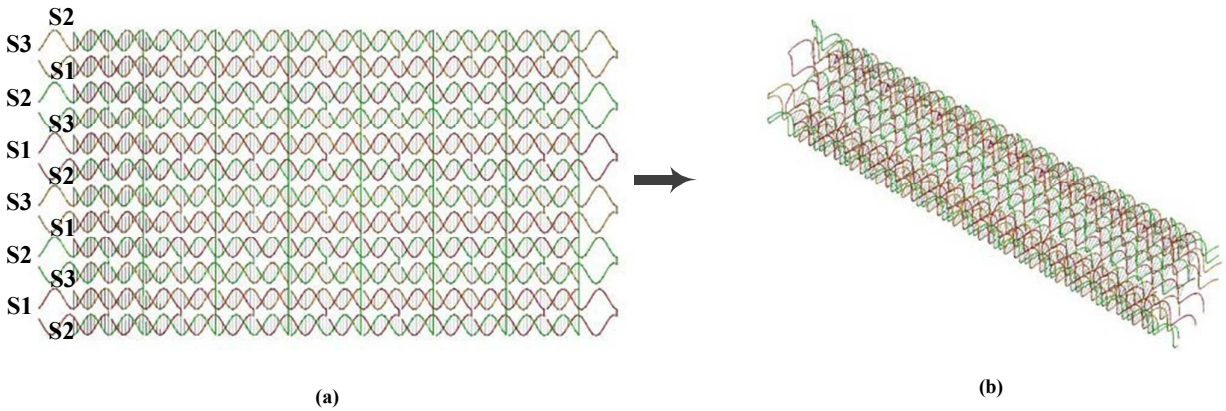


Figure 2. Secondary structures **(a)** The accumulation of intrinsic curvature of brick SS and the cyclization of the assembled lattice. **(b)** DNA nanotube

During the execution of specified program through one-pot annealing, the specified complementary relationship between the domains of brick SSTs will direct DNA molecules toward a thermodynamic minimum on the free energy landscape [29], by which the desired DNA nanoribbon will be assembled. The DNA nanoribbon could be generalized by programming the formation of n-helix ribbons, using brick SSTs (S1, S2, and S3) and manipulative strands (S1-1, S1-2 and S1-3) on the bottom edge, see Figure 3 (a) and (b). The presence of the manipulative strands on the bottom edge will avoid the complementation of two edges of the tubes, forcing the DNA nanotubes to open into ribbons, shown in Figure 3(c).

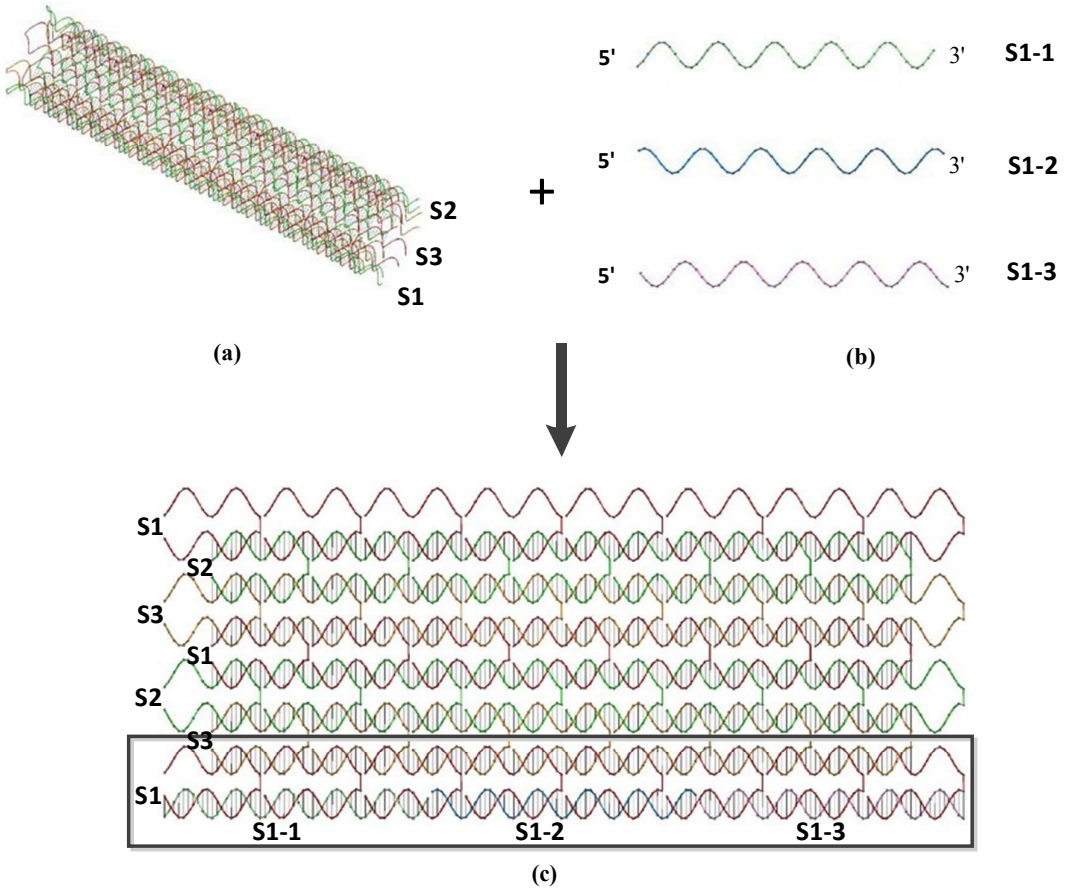


Figure 3. The formation of DNA nanoribbon **(a)** Nanotube formation with SSTs, S1, S2 and S3. **(b)** Manipulative strands on the bottom edge. **(c)** Nanotubes opened into ribbons with the presence of the manipulative strands according to the thermodynamic minimum on the free energy landscape.

To design manipulative strands, it is firstly divides domains 1 and 2 of S1 into 3 parts evenly, labeled by domain 11, domain 12 and domain 13. Domains 1 and 2 of S1, shown in Figure 1, have 21 nucleotides, so each of domain 11, domain 12 and domain 13 is composed of 7 nucleotides, see Figure 4.

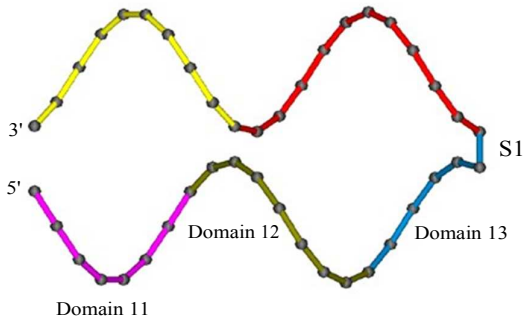


Figure 4. Dividing domains 1 and 2 of SST S1 into three domains 11, 12 and 13 evenly

Manipulative strand S1-1 is complementary to DNA strand concatenating sequentially domain 12, domain 11, domain 13, domain 12, domain 11, domain 13, domain 12 and domain 11 (from 3' to 5' end); manipulative strand S1-2 is complementary to DNA strand concatenating sequentially domain 11, domain 13, domain 12, domain 11, domain 13, domain 12, domain 11 and domain 13 (from 3' to 5' end); and manipulative strand S1-3 complementary to DNA strand concatenating sequentially domain 13, domain 12, domain 11, domain 13, domain 12, domain 11, domain 13 and domain 12 (from 3' to 5' end). In this way, manipulative strands S1-1, S1-2 and S1-3 (from 3' to 5' end) can be represented by DNA strand concatenating sequentially complementary domain of domains 11, 12 and 13. These manipulative strands are shown in Figure 5, where domain 11', domain 12' and domain 13' represent complementary strand of domain 11, domain 12 and domain 13, respectively.

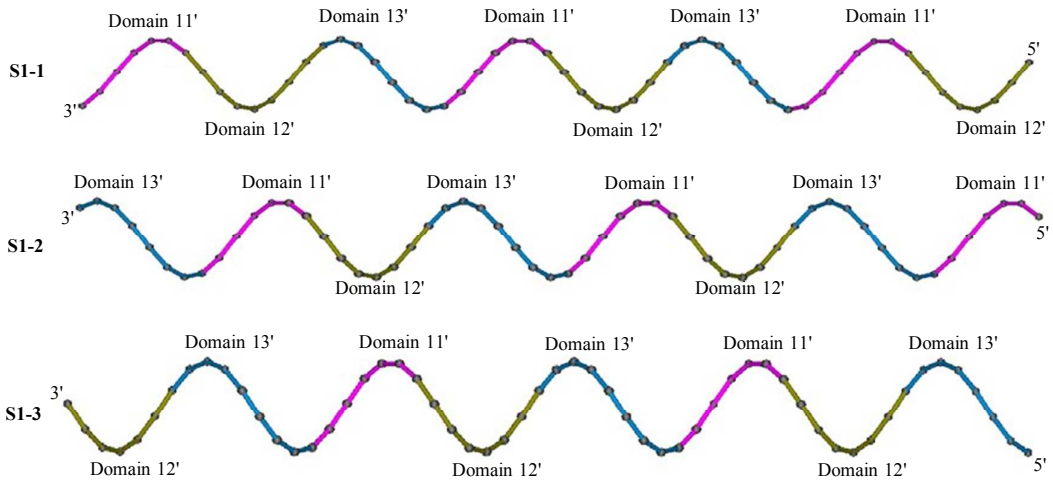


Figure 5. Manipulative strands S1-1, S1-2 and S1-3 (from 3' to 5' end)

It is easy to find out that each manipulative strand will have 56 nucleotides (8 domains with each domain having 7 nucleotides). We can check that it needs 8 brick SST S1 to fully complement the stand by concatenating manipulative strands S1-1, S1-2 and S1-3. Manipulative strands S1-1, S1-2 and S1-3 can complement the edge DNA strand S1, thus avoiding the formation of DNA nanotubes. In this way, DNA nanotubes will be "opened" to form DNA nanoribbons. The complementary relationship between strand S1 and manipulative strands (S1-1, S1-2 and S1-3) can be found in Figure 3 (c).

Mathematically, the length of the ribbon should be the value of least common multiple between lengths of SST S1 and the manipulative strand. Therefore, it is achieved a strategy to control the length of DNA nanoribbon by designing SSTs with different lengths.

Packed strands with labels le13, le32, le21, re13, re32 and re21 are designed and used to avoid the nanoribbon's horizontal generation from both left and right sides. The general functions of strands le13, le32, le21, re13, re32 and re21are indicated in Figure 6.

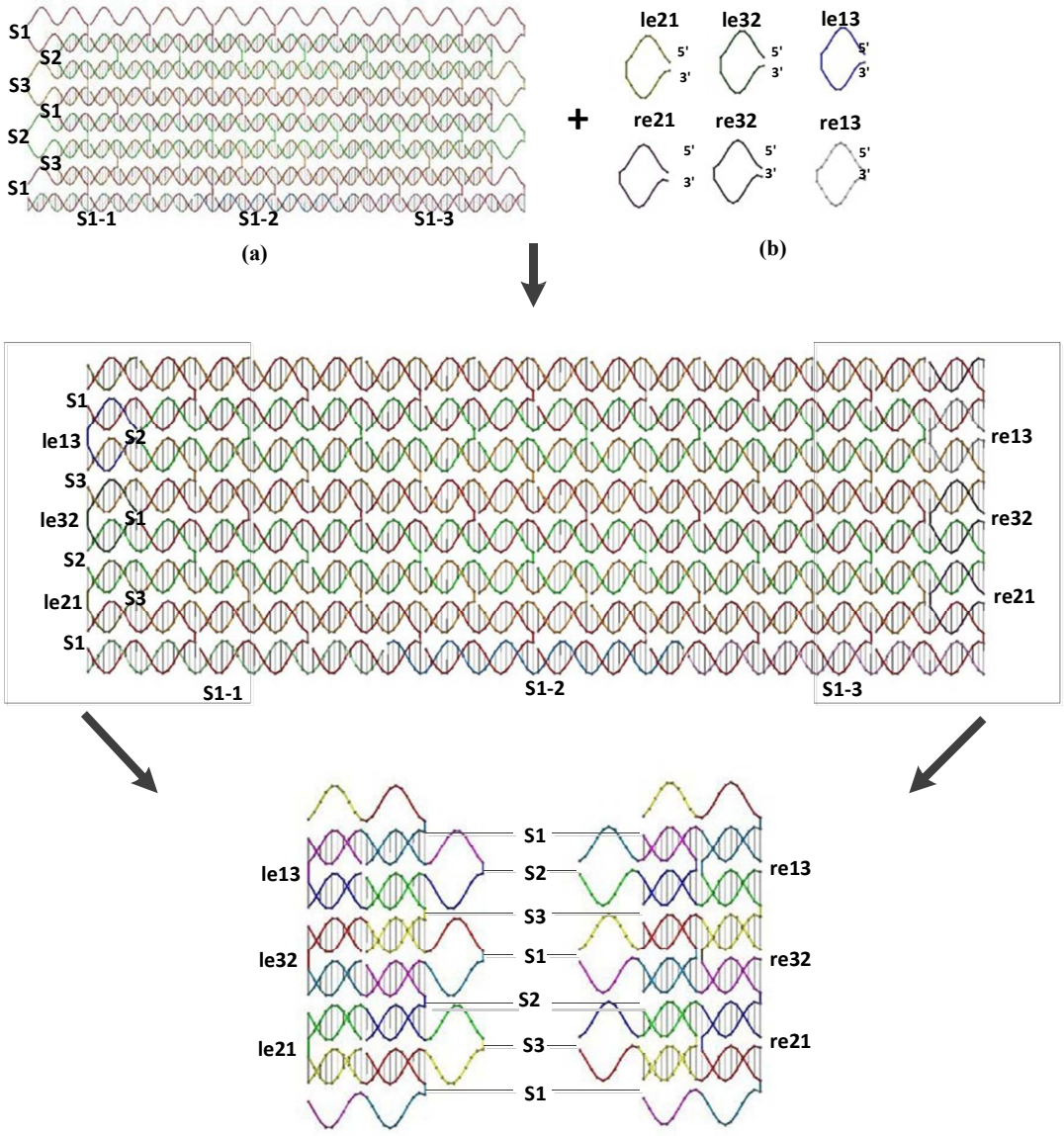


Figure 6. The general functions of strands le13, le32, le21, re13, re32 and re21, where domains with same color indicate that they are complementary.

Strands le13, le32, le21, re13, re32 and re21 force the horizontal extension nanoribbons. Each strand consists of 21 nucleotides, and each of the following pairs of domains is complete complementary: domain 1 of le13 and domain 1 of S1, domain 2

of le13 and domain 4 of S3, domain 1 of le32 and domain 1 of S3, domain 2 of le32 and domain 4 of S2, domain 1 of le21 and domain 1 of S2, domain 2 of le21 and domain 4 of S1, domain 1 of re13 and domain 2 of S1, domain 2 of re13 and domain 3 of S3, domain 1 of re32 and domain 2 of S3, domain 2 of re32 and domain 3 of S2, domain 1 of re21 and domain 2 of S2, domain 2 of re21 and domain 3 of S1. By regulating the concentration ratio between the set of manipulative strands and the packed strands, DNA nanoribbons with variable lengths can be obtained. In addition, the proportion of long nanoribbons decreases with the increase of the concentration ratio between the set of manipulative strands and the packed strands.

3. Experimental Results

The oligonucleotides used in our experiments were acquired from Sangon Biotech Company with PAGE purification. The brick SSTs, S1, S2 and S3 (whose sequences structure are shown in supplementary), are used to constructed nanotubes and nanorings by one-pot annealing from 94ºC to room temperature in TAE buffer (10mM Mg2＋, 20mM Tris with pH 7.6－8.0, 2mM EDTA) over 20 hours with a final concentration of 1μM for each strand. The ratio between three SST strands S1, S2 and S3 are the same, that is, 1:1:1. After annealing, the sample was kept in a refrigerator at 4ºC before being examined by AFM. The results are shown in Figure 7.

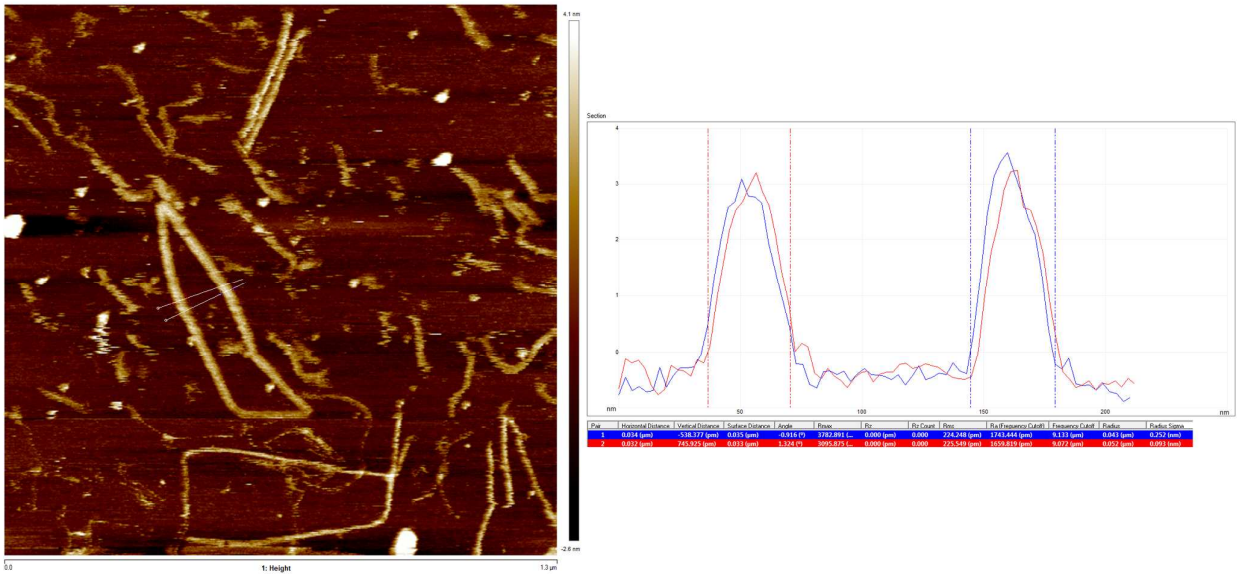
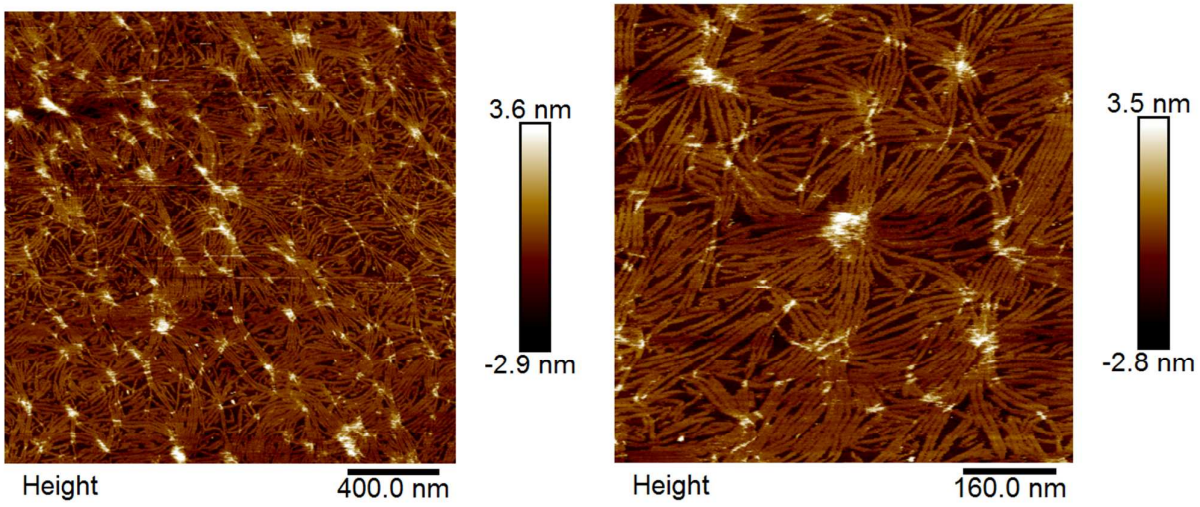


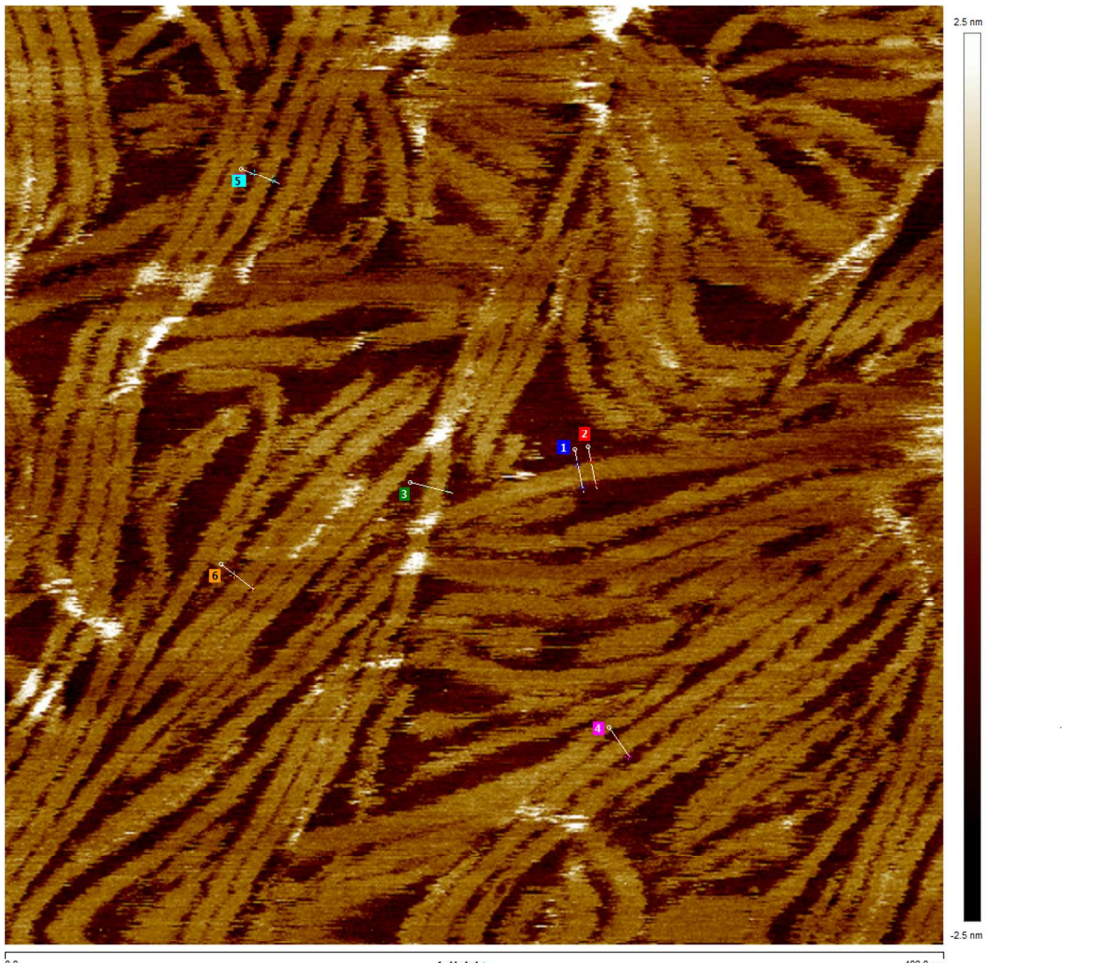
Figure 7. AFM images of DNA nanotubes and nanorings assembled by SSTs S1, S2 and S3

In left part of Figure 7, it shows the AFM image of the nanotubes and nanorings with 1.3μm×1.3μm scan size, where the data height range is -2.5－4.0nm; in right part of Figure 7, section date of the AFM picture is given, the height of the structure is about 3.5nm and the width is about ~34nm, which indicated the DNA nanostructure that form

from the three bricks SSTs, S1, S2 and S3 also curves into nanotubes as Yin Peng's works of programmable nanotubes with unique SST strands [30], or even nanorings. The normal width ~34nm of the nanoring suggested the continuous assemble of strands S1, S2 and S3 to about 15 helix for one layer considering the gap between two helices. The wide distribution of the width of nanotubes may be due to the random curve during the repeating assembling of the three bricks.

After that, manipulative strands S1-1, S1-2 and S1-3 (the sequences were shown in supplementary) are added to "open" the assembled DNA nanotubes and to lead the formation of DNA nanoribbons. All the three bricks SSTs strands and manipulative strands in this sample are in a concentration ratio of 8:3. The final concentration of oligonucleotide mixture were 0.5 μM. The mixture is annealed from 94°C to room temperature in TAE buffer (10mM Mg2+, 20mM Tris with pH 7.6－8.0, 2mM EDTA) over 20 hours with one-pot strategy. The sample is kept in a refrigerator at 4ºC before being examined by AFM, shown in Figure 8.





Pair	Horizontal Distance	Vertical Distance	Surface Distance	Angle	Rmax	Rz	Rz Count	Rms	Ra (Frequency Cut...	Frequency Cutoff	Radius	Radius Sigma
1	9.613 (nm)	126.758 (pm)	11.488 (nm)	0.755 (°)	1977.307 (pm)	0.000 (pm)	0.000	742.248 (pm)	667.007 (pm)	0.048 (nm)	6.993 (nm)	0.319 (nm)
2	9.613 (nm)	-160.948 (pm)	11.083 (nm)	-0.959 (°)	1890.086 (pm)	0.000 (pm)	0.000	570.341 (pm)	603.767 (pm)	0.048 (nm)	7.536 (nm)	0.275 (nm)
3	8.899 (nm)	-541.222 (pm)	9.728 (nm)	-3.480 (°)	1875.068 (pm)	0.000 (pm)	0.000	443.275 (pm)	680.303 (pm)	0.048 (nm)	7.172 (nm)	0.139 (nm)
4	8.623 (nm)	108.754 (pm)	9.578 (nm)	0.723 (°)	1562.671 (pm)	0.000 (pm)	0.000	752.004 (pm)	593.065 (pm)	0.052 (nm)	6.508 (nm)	0.127 (nm)
5	8.516 (nm)	112.209 (pm)	9.578 (nm)	0.755 (°)	1824.961 (pm)	1824.961 (pm)	2.000	679.070 (pm)	746.160 (pm)	0.098 (nm)	6.335 (nm)	0.201 (nm)
6	9.917 (nm)	461.021 (pm)	10.673 (nm)	2.662 (°)	1474.397 (pm)	1474.397 (pm)	2.000	449.403 (pm)	456.208 (pm)	0.050 (nm)	8.892 (nm)	0.175 (nm)

Figure 8. AFM images of DNA nanoribbon assembled by brick SSTs S1, S2 and S3 and manipulative strand S1-1, S1-2 and S1-3.

It is shown in the top of Figure 8 the AFM image of the nanoribbon with 2 μm×2 μm scan size; the height is from -2.9 to 3.6 nm and a zoom in AFM image with 800 nm×800μm scan size to show the details of nanoribbon. The result indicates

that the manipulative strands S1-1, S1-2 and S1-3 can prevent the random curve of the three bricks SSTs, S1, S2 and S3 into nanotubes and make the formation of long ribbons with regular width successfully.

In the bottom, a zoom in AFM image of the nanoribbon with 400 nm×400 nm scan size and it's section data is shown, where the data height range is -2.5－2.5 nm. It is shown by the section data of marker pairs 1 to 6 that the height of the ribbon is about 1.3nm, and the mean of surface distance of two markers (width of nanoribbons) is about 10.35 nm and standard deviation is 0.84. The average length of ribbons is over 800 nm.

We add packed strands le13, le32, le21, re13, re32 and re21 (whose sequences are shown in supplementary) into the mixture, by which we can control the length of the DNA nanoribbon by regulating the concentration ratio between manipulative strands and packed strands. Three concentration ratios are included in our experiments, which are 1:1, 1:2 and 1:4 of manipulative strands and packed strands respectively, in all the samples the three brick strands kept the same concentration ratio with manipulative strands as 8:3 as previous samples shown in Figure 8. The samples are annealed with one-pot strategies from 94ºC to room temperature in TAE buffer (10mM Mg2＋, 20mM Tris with pH 7.6－8.0, 2mM EDTA) over 20 hours. After annealing, they are kept in a refrigerator at 4ºC before being examined by AFM. The AFM images and the pie chart are shown in Figure 9.

The samples are observed by AFM in fluid-scanasyst mode. A drop of 1 μl sample solution is pipetted onto a freshly cleaved mica surface and kept for 2 min for adsorption; the Mg2+ in the buffer acted as counter ions to allow the negatively charged DNA to bind to the negatively charged mica surface. Two portions of 15 μl buffer are added onto the mica and AFM tip, respectively. AFM images are obtained after adjusting involved parameters.

In Figure 9, the AFM images of DNA nanoribbon assembled with 1:1, 2:1 and 4:1 concentration ratio between manipulative strands and packed strands are shown, where the pie chart is the packed strands' distribution in DNA nanoribbon with different length. The average width of these ribbons is about 15 nm, 16nm and 26 nm with respect to concentration ratio 1:1, 2:1 and 4:1 between manipulative strands and packed strands. From packed strands' distributions in DNA nanoribbon with different lengths, it is concluded that the length of DNA nanoribbons decreases with the increase of the concentration ratio between the set of manipulative strands and the packed strands.

Therefore, experimental observations suggest the feasibility of our method to assemble size controllable DNA nanoribbons with three types of reusable brick

SSTs, where the control of size is achieved by regulating concentration ratio between manipulative strands and packed strands. It is concluded that the proportion of DNA nanoribbons decreases with the increase of the concentration ratio between manipulative strands and packed strands.

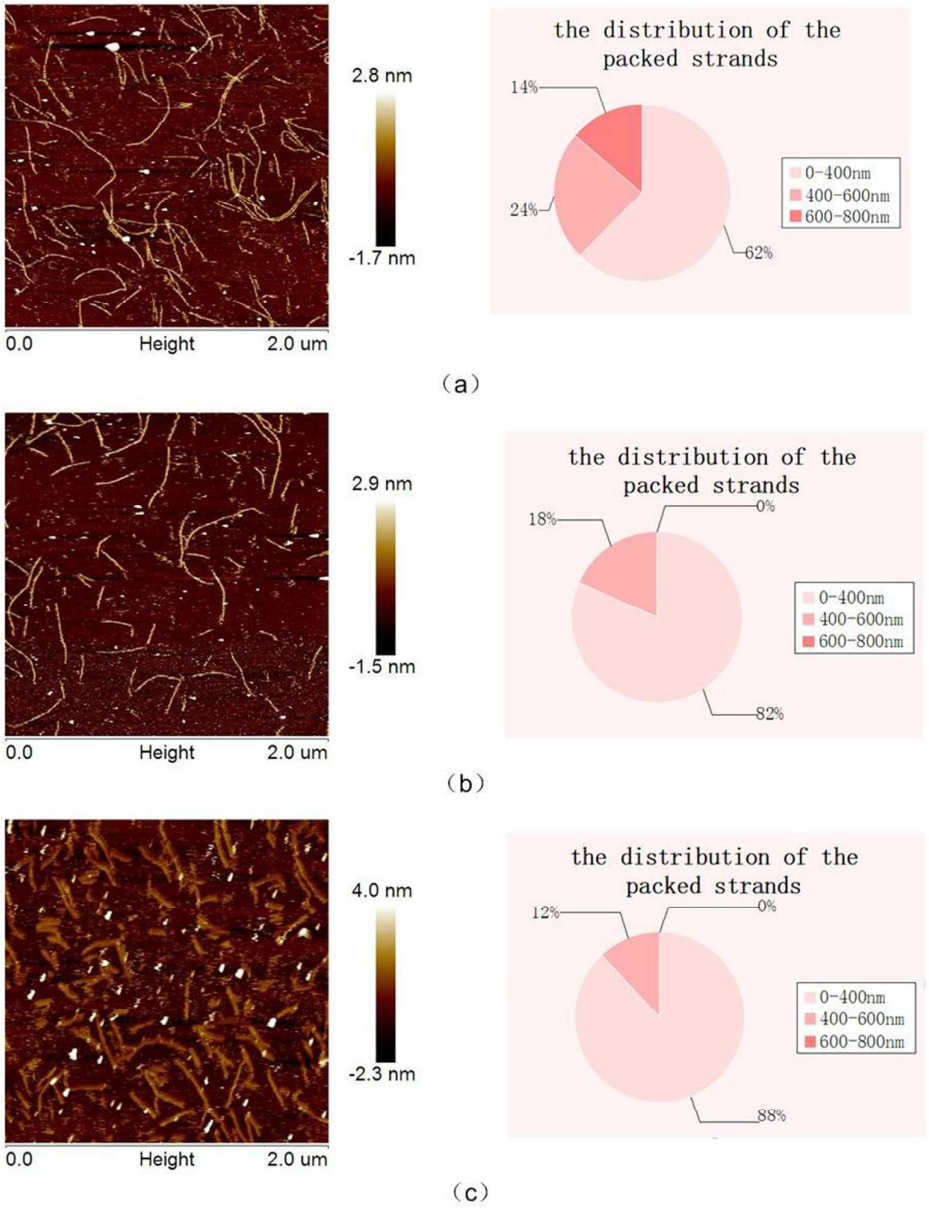


Figure 9. AFM images of DNA nanoribbon with different length

4. Conclusion and Final Remarks

In this work, we propose a novel approach to assemble DNA nanoribbons with controllable size by three types of reusable brick SSTs, where the control of ribbon size is achieved by regulating the concentration ratio between s manipulative strands and packed strands. It is worthy to point out that three types of brick SSTs are sufficient in assembling DNA nanoribbons with different sizes by our method, thus achieving a universal and low cost method.

With the presence of intrinsic curvature of SST, more than 15-helix with repeating three SST strands will be randomly folded into nanotubes with wide distribution, and the height of nanotubes is about 3.5nm. With long manipulation strands which make a botoom boundary, the three repeating SST strands assembled into DNA nanoribbons with regular width about 9.5nm. It is obtained by AFM that the height of the ribbon is about 1.3nm, and the regulation of the width of nanoribbon must due to the guildence of the long manipulation strands during the self-assembly of three SST strands. Brownian motion and thermodynamic minimum on the free energy landscape impacted the growing trend of ribbon. Therefore, the length of nanoribbons is multiples of the length of 16 helical turn of dsDNA.

It is obtained that the average length of ribbons is over 800 nm. Three groups of experiments with different concentration ratios between manipulative strands and packed strands are operated. It is concluded that the proportion of DNA nanoribbons decreases with the increase of the concentration ratio. Moreover, high concentration of packed strands can prevent the growth of ribbon length, thus increasing slightly the width of nanoribbon. The average width of ribbons is about 15 nm, 16 nm and 26 nm with the concentration ratio 1:1, 2:1 and 4:1 respectively. The strategy used here is based on the structural flexibility of SST, which provides a way to regulating size of DNA nanostructures by the ratio of involved concentrations of strands. It gives a new research topic in DNA nanotechnology.

There are many related topic deserve further research. For example, by precise controlling the concentration of strands, further control over the size of ribbons may be achieved. In addition, using more boundary strands may lead to nanostructures of other shapes, such as triangles and finite long rectangles. Also, the ribbon with finite dimensions constructed here are likely to find applications ranging from biophysics to electronics. In biophysics, the programmable sizes of the ribbon make it attractive synthetic model system. In electronics, the metallized nanoribbons from [30] may result in conductive metallic nanowires with controlled length and, hence, controlled electronic properties.

5. Acknowledgement

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